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Abstract

A status report on the preparation and properties of $Ga(As_{1-x}P_x)$ p-n junction lasers is presented. Halogen vapor transport synthesis of $Ga(As_{1-x}P_x)$ and its preparation into laser junctions are described. Electrical and optical properties of $Ga(As_{1-x}P_x)$ laser junctions are discussed. The present limitations in these properties are related to material problems and the very early state of development of $Ga(As_{1-x}P_x)$, and are discussed in this context.

Introduction

In this report we wish to describe briefly some of the salient features of now approximately five or more months experience with the preparation and properties of $Ga(As_{1-x}P_x)$ p-n junction lasers. Because much data so far available are incomplete, some of the results at this point must be considered tentative and subject to revision as work on $Ga(As_{1-x}P_x)$ progresses.

Although the electrical, optical, and device properties of $Ga(As_{1-x}P_x)$ junction lasers are understandably of great interest, the work to date points out that by far currently the greatest problems and those deserving first attention are those involving the growth, doping, and perfection of $Ga(As_{1-x}P_x)$ crystals. Hence, even though the laser junctions prepared in this material are of importance in their own right, we have considered them at this point mainly a means to study $Ga(As_{1-x}P_x)$. It can be concluded that the laser junctions now available are very much limited by the deficiencies of $Ga(As_{1-x}P_x)$ crystals and can not be fairly compared with GaAs p-n junction lasers until $Ga(As_{1-x}P_x)$ crystal growth has been more extensively developed. The basis for these statements will become evident in the following sections which describe $Ga(As_{1-x}P_x)$ crystal and junction preparation and junction properties.

A. Preparation of $Ga(As_{1-x}P_x)$ Crystals and Laser p-n Junctions

As previously mentioned[1], we have found it convenient to employ the halogen vapor transport process[2] for synthesizing $Ga(As_{1-x}P_x)$ crystals for use in laser junctions. In the present work commercially available GaAs and GaP separately prepared by halogen vapor transport[2] are sealed in a quartz vessel with a quantity of column VI donor and a small quantity of metal halide which supplies the halogen for transport. The sealed ampoule is heated at a temperature in the range from 1000°C to 1100°C at the position of the source GaAs and GaP. The GaAs and GaP is simultaneously vapor transported and deposited as $Ga(As_{1-x}P_x)$ at one end of the ampoule held at a lower temperature (between 900°C and 1050°C).

With this process we have been able to grow some large single crystal sections and a number of polycrystalline ingots of large crystallite diameters, which have been further processed into laser junctions by methods more or less similar to those employed with GaAs. Of the crystals so prepared a small quantity has yielded material of "laser quality". $Ga(As_{1-x}P_x)$ prepared from commercially available GaP thus far has failed to yield laser junctions, perhaps because of carbon contamination. In a number of cases "good quality" $Ga(As_{1-x}P_x)$ which has provided strong incoherent junction luminescent light sources has failed to "lase" because of insufficient donor doping in the crystal. The crystals which have "lased" have been doped to $\sim 10^{18}$ donors/cm^3 and have been freer of strain than "non-lasing" crystals[3]. Also, the crystals that "lase" have tended to be freer of various dark spots which are evident under IR examination[3].

The fact that preparation of $Ga(As_{1-x}P_x)$ of adequate quality poses the greatest obstacle to preparation of laser junctions is evident from a cursory examination of the journal literature. Lasers in $Ga(As_{1-x}P_x)$ were reported[1] only slightly after the first

reported GaAs junction lasers of Hall, et al[4], and well before most subsequent confirming reports of laser action in GaAs. Nevertheless, recent reports and, in fact, the journal literature for a period well beyond five months of Hall's, et al[4] first report of GaAs junction lasers contain hardly any mention of $Ga(As_{1-x}P_x)$, almost as though (contrary to fact) $Ga(As_{1-x}P_x)$ will not "lase". This queer circumstance, in spite of the great interest, flexibility, and importance of $Ga(As_{1-x}P_x)$, is due simply to the lack of laser quality $Ga(As_{1-x}P_x)$ and the difficulty in its synthesis to requisite perfection, purity, and doping.

Following synthesis of "suitable" $Ga(As_{1-x}P_x)$ crystals the procedures we have used for fabrication of laser junctions follow closely those employed with GaAs. Suitably cut wafers are polished and are zinc diffused in sealed ampoules at temperatures as high as 1000°C. Long strips are cut out of the wafers, polished on the cut sides at right angles to the junction plane, and are separated into small sections to which electrical connections can be easily alloyed. (Or, when desired, p+ zinc-doped $Ga(As_{1-x}P_x)$ layers have been epitaxially grown by the vapor transport process[2] on n-type wafers to form p-n junctions.)

From the crystals grown to date of the order of 100 laser p-n junctions have been assembled. It is interesting to note that since these laser junctions have operated at a number of wavelengths, almost all different, $Ga(As_{1-x}P_x)$ has perhaps already "lased" at as many different wavelengths as <u>all</u> other solid state lasers combined. This could be expected[1], for as shown by Fig. 1, reproduced from Ehrenreich's work[5] and changed to correspond to 77°K, $Ga(As_{1-x}P_x)$ is expected to be direct over a composition range from 0 % to 50 % phosphorus ($0 \leq x \leq \sim 0.5$) and a corresponding wavelength range from ~8400Å to ~6000Å (or less, 77°K).

It should be mentioned that our experimental data and points when inserted upon Fig. 1 tend to fall below the theoretical curve

and indicate an energy gap less than predicted. This may imply that heavy donor or acceptor doping in our junctions smears the band edges and narrows the energy gap. This would imply further that it may be difficult or impossible to achieve laser wavelengths near 6000Å in $Ga(As_{1-x}P_x)$. Also, if this interpretation is correct, it may be equally difficult or impossible to establish that discrete donor or acceptor levels are involved in the light producing recombination process. In fact, in contradiction to several schools of thought believing donor or acceptor levels are involved in the recombination luminescence process in GaAs, we see no evidence in $Ga(As_{1-x}P_x)$ to favor this belief at this time.

B. Electrical and Optical Properties of $Ga(As_{1-x}P_x)$ Laser Junctions

In an earlier report[6] we described the high quality I-V characteristics and sharp avalanches which can be obtained in $Ga(As_{1-x}P_x)$ p-n junctions. Recent investigations have shown that in several respects the avalanche properties of $Ga(As_{1-x}P_x)$ may excel silicon[7]. On units of comparable area lower noise and lower dynamic impedance have been observed in $Ga(As_{1-x}P_x)$.

Aside from various inhomogeneities in $Ga(As_{1-x}P_x)$, currently one of the more serious problems is that associated with contaminating impurities and resultant deep levels. The effects of deep levels on $Ga(As_{1-x}P_x)$ p-n junctions are generally evident at low temperatures. Where such levels abound, just as in GaAs the forward I-V characteristic becomes higher impedance at lower temperatures, and may even exhibit negative resistance behavior. These are familiar double-injection effects[8] and are a direct consequence of deep levels. Unfortunately these effects compete with the high-efficiency, direct recombination process by allowing carrier recombination to occur in "steps" via deep levels. In the many instances that we have observed these effects, as expected, we have not been able to obtain stimulated emission. So far, we have not identified specific impurities which contribute deep levels but regard O_2 as a likely suspect since it is a major constituent of our synthesis vessel and since a number of the elements present in the synthesis process may react with the quartz synthesis vessel. At any rate, the observation of deep level effects in $Ga(As_{1-x}P_x)$ junctions underscores the need to continue improving synthesis and growth techniques and crystal quality.

An interesting and important question concerning laser junctions is that of the mechanism of light generation and the

related question of where in the junction region the recombination process (and radiation) occur. To study this problem we have examined ~ 10 laser junctions which have been cross-sectioned and polished normal to the junction plane. With a mild etch slight "moats" were cut into the polished sections in order to reveal the junction transition regions. In order to remove errors of focusing inherent in dispersion effects at different wavelengths, we examined the junction cross-sections at high power under incident red light and with the sectioned but operative junctions supplying recombination radiation (red).

In each case in which the experiment was carefully performed the source of recombination radiation (red) and junction "moat" coincided, with no indication that either n-side or p-side of the junction transition supplied more light. Assuming that the etched moat truly coincides with the junction transition region, which is a reasonable first order assumption, we conclude (until better evidence is obtained) that the recombination process (and radiation) occurs right in the junction transition region. These results and the fact that these junctions are heavily doped on n- and p-type sides suggest that these devices are much like p-i-n structures of exceedingly narrow extent between n- and p-side (very narrow "i"-region, but not "tunneling" narrow or a true "step" transition). This conclusion is consistent with the earlier observation of the "snap-off" behavior of stored charge in $Ga(As_{1-x}P_x)$ laser junctions[6], again indicative of the fact that the injected positive and negative charge is in the form of a <u>thin</u> sheet probably residing right in the transition or thin "i" region.

It should be noted that the discussion above does not necessarily imply that discrete acceptor or donor levels are not involved in the recombination process. However, the evidence that

the recombination radiation originates in the junction transition region (rather than on either side) plus the likelihood that the high acceptor and donor concentrations smear both band edges through the transition region make uncertain the view that discrete levels play a primary role in the recombination process.

In an earlier publication[9] we showed that at the current threshold for laser action $Ga(As_{1-x}P_x)$ junctions first exhibit laser action in discrete regions through the junction plane from one polished face to the other. As the driving current is increased these lasing regions increase in size or other separate regions begin to exhibit laser action, and not necessarily at the same or mode-related wavelength of the first region. From cross-sections of laser junctions we have found that some junctions do not lase uniformly throughout the junction plane because of pronounced localized dips or spikes in the junction plane. In fact, usually in $Ga(As_{1-x}P_x)$ junctions polished on all four sides, i.e. polished on the four sides bounding and at right angles to the junction plane, laser action will generally occur anomalously along the short dimension of the junction plane rather than along the long dimension because of localized dips, spikes, and assorted flaws in the junction. Laser action occurs in the short dimension because light traveling from polished face to polished face in the junction plane is not as apt to find an interruption in geometry as in the long dimension. These observations further point up the need for an intensive effort to improve the quality of $Ga(As_{1-x}P_x)$ crystals.

The usual optical transmission procedures for examining the band gap of $Ga(As_{1-x}P_x)$ prepared by halogen vapor transport generally show that the composition of the material is close to that predicted on the basis of the known initial quantities of GaAs and GaP used in the synthesis process. Laser junctions, on the

contrary, show that even though $Ga(As_{1-x}P_x)$ can have a well-defined band structure and a good microscopic composition, significant macroscopic fluctuation can occur in As:P ratio in the material. This is evident from the laser junction spectra shown in Fig. 2. The sharp transition from incoherent operation at 20A to laser operation at 22A is shown by curves a) and b), both taken with spectrometer and amplifier settings constant. Curve b) shows laser action at wavelengths separated by almost 40Å and an independently measured spatial separation of $\sim 2 \times 10^{-2}$ cm between the two laser beams. The diode operates almost as though it were two different laser junctions, each with the same length cavity (Fabry-Perot structure) but with differing $Ga(As_{1-x}P_x)$ composition and band gap. In laser junctions in which the As:P ratio is spatially more homogeneous across the junction plane the spectral output is confined to a much smaller band than the diode of Fig. 2. Also, where a number of spectral peaks are observed they occur as well-defined modes with uniform spacing. In one such case we have counted 30 uniformly spaced modes in a spectral band $\sim$ 30Å wide.

An indication of the efficacy of laser action in $Ga(As_{1-x}P_x)$ p-n junctions is supplied by Fig. 3, which shows the spectral output of junction 95-1 with current excitation [a) 12A] just below threshold and [b) 13A] just above threshold (spectrometer, photomultiplier voltage, and amplifier settings all constant). Above threshold the spectral output narrows sharply, and for less than a 10 % change in current drive (excitation) the peak spectral output increases by more than 10x. Laser junctions of this type with less than 2 mW average input power produce enough coherent light (half the output coherent light) so that the diverging beam can be condensed, and focused and be easily observed on screens at distances beyond 60 feet. For the diodes we have examined the coherent light output falls in a cone of esti-

mated half-angle $\sim 5^{\circ}$. The coherent light output can be displayed easily on a screen or ground glass plate in a red-colored version of the familiar diffraction pattern first described by Hall, et al[4].

Contributors and Contract Activity

Scientists and technical staff who contributed to the work reported:

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C. V. Bielan	F. A. Carranti
	B. G. Hess
	S. J. Lubowski

Papers published under contract:

1. N. Holonyak, Jr. and S. F. Bevacqua, Appl. Phys. Letters 1, 82 (1962).
2. N. Holonyak, Jr., et al., Proc. Inst. Elec. Elec. Engrs. 51, 364 (1963).
3. N. Holonyak, Jr., Electronics 36, 35 (March 1, 1963).

Seminar presentations during contract:

N. Holonyak, Jr., Univ. of Illinois, Dec., 1962.

N. Holonyak, Jr., Syracuse Univ., Jan., 1963.

N. Holonyak, Jr., Syracuse Section I.E.E.E.-P.G.M.T.T., Feb., 1963.

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1. N. Holonyak, Jr. and S. F. Bevacqua, Appl. Phys. Letters 1, 82 (1962).

2. N. Holonyak, Jr., D. C. Jillson, and S. F. Bevacqua, Chap. 15, pp. 49-59, Metallurgy of Semiconductor Materials 15 (Proceedings of A.I.M.E. Conf., Los Angeles, Aug. 30-Sept. 1, 1961; Interscience Div. of John Wiley, N.Y., N.Y., 1962).

3. We are grateful to R. N. Hall for IR and birefringence analysis of a number of our crystals and laser junctions.

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7. F. Gentry, private communication.

8. N. Holonyak, Jr., Proc. Inst. Radio Engrs. 50, 2421 (1962).

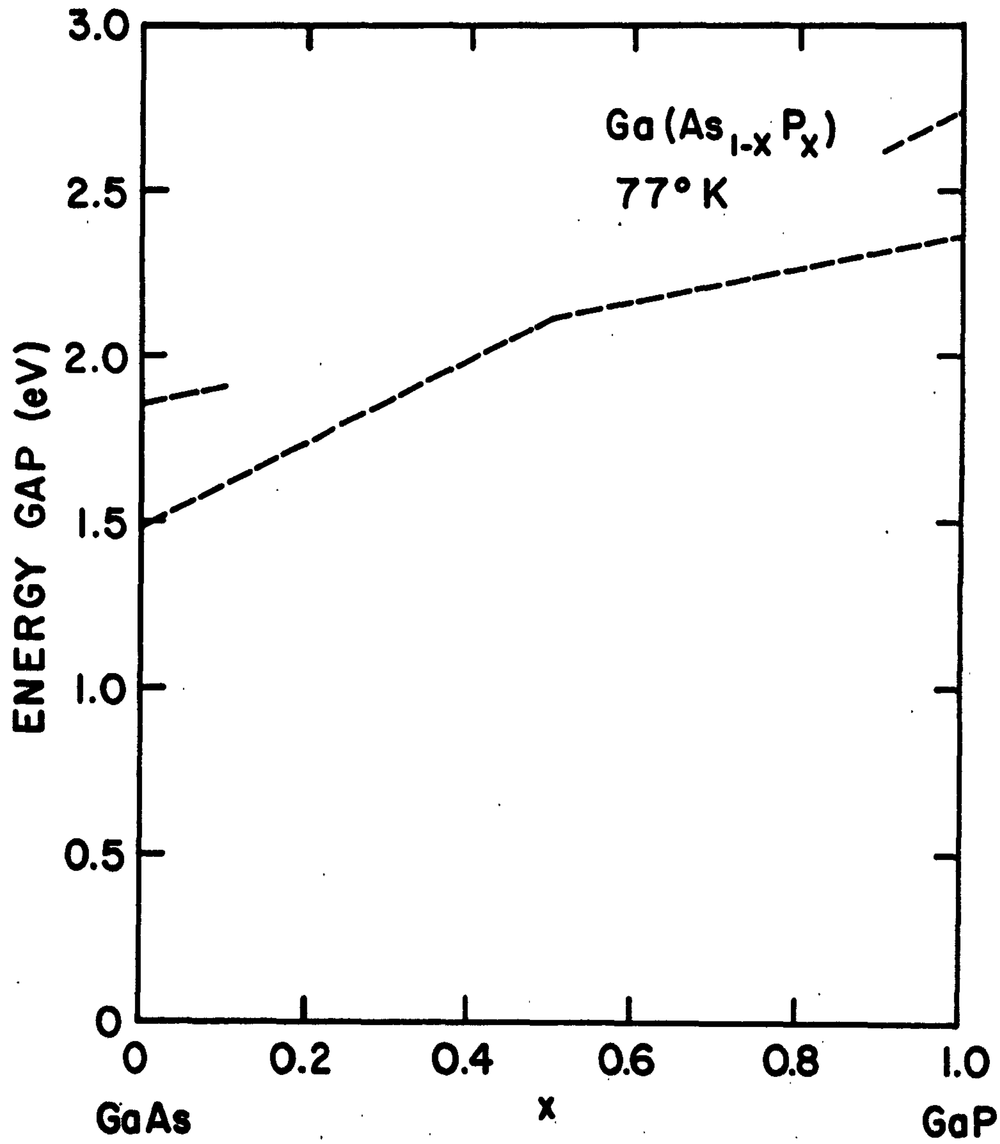
9. N. Holonyak, Jr., Electronics 36, 35 (March 1, 1963).

Figure Captions

Fig. 1 Energy gap as a function of composition for $Ga(As_{1-x}P_x)$ at 77°K (after Ehrenreich, J. Appl. Phys. 32, 2155, 1961).

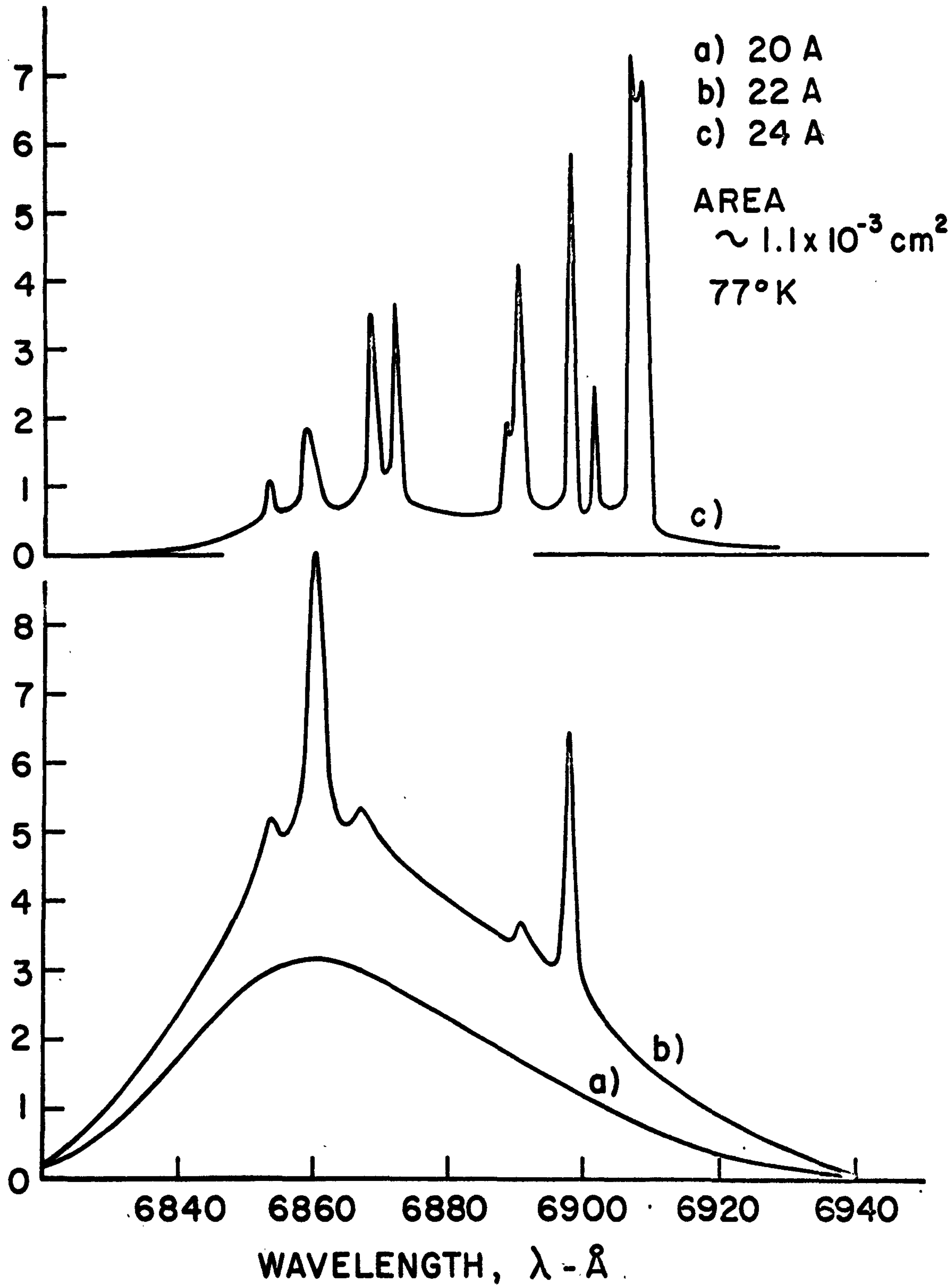
Fig. 2 Spectral output of $Ga(As_{1-x}P_x)$ diode 95-4 at 77°K. a) Below threshold (20A), b) above threshold (22A). At 22A output consists of two coherent light sources separated spatially by $\sim 2 \times 10^{-2}$ cm and differing in wavelength by ~ 40Å.

Fig. 3 Spectral output of $Ga(As_{1-x}P_x)$ diode 95-1 at 77°K when operated just below threshold a) and just above threshold b). Scale same for a) and b).



3.0
2.5
2.0
1.5
1.0
0.5
0
ENERGY GAP (eV)
Ga(As1-x Px)
77° K
0
0.2
0.4
0.6
0.8
1.0
GaAs
x
GaP

FIG. 1



a) 20 A
b) 22 A
c) 24 A
AREA
~ 1.1 x 10^{-3} cm^2
77° K
c)
b)
a)
0
1
2
3
4
5
6
7
8
6840
6860
6880
6900
6920
6940
WAVELENGTH, λ - Å

FIG. 2

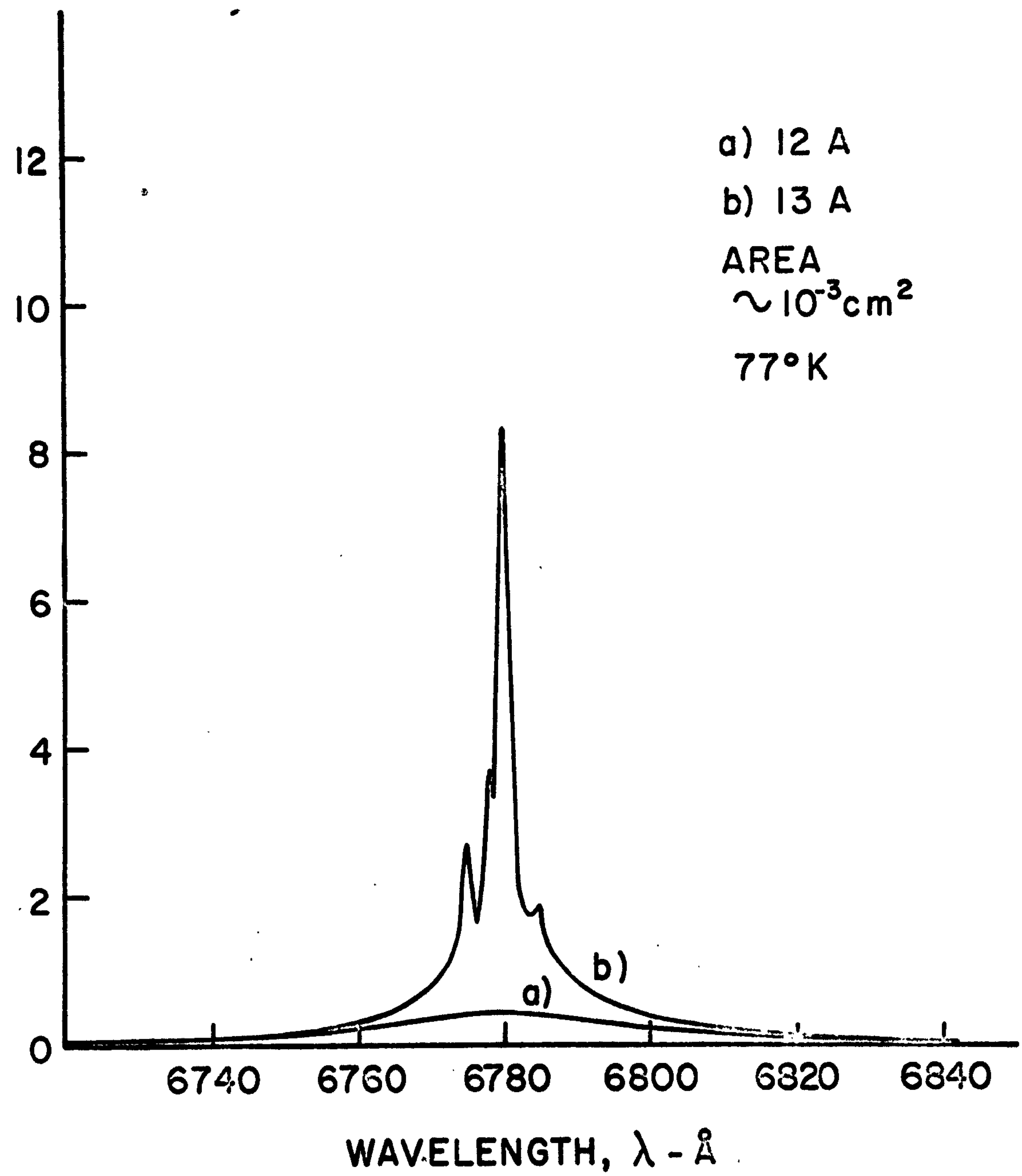

a) 12 A
b) 13 A
AREA
~10⁻³cm²
77°K
12
10
8
6
4
2
0
6740
6760
6780
6800
6820
6840
WAVELENGTH, λ - Å
a)
b)

FIG. 3

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